



Removal of lead and zinc from battery industry wastewater using electrocoagulation process: Influence of direct and alternating current by using iron and stainless steel rod electrodes



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ABSTRACT

In this study the efficiency of electrocoagulation by direct and alternating current in the removal of lead and zinc has been evaluated. Wastewater samples were taken from the battery building industry. In the electrochemical cell, metal iron and stainless steel rod electrodes were used and were connected together serially and monopolar to the power source. By using alternating current the removal of lead and zinc was done successfully with iron electrodes and in the current density of 6 mA/cm² which was respectively 96.7% and 95.2% and with stainless steel electrodes in current density of 8 mA/cm² was 93.8% and 93.3%. By using direct current the optimum removal of lead and zinc with iron electrodes was respectively with 97.2% and 95.5% in current density of 6 mA/cm² and with stainless steel electrodes was equal to 93.2% and 92.5% in current density of 8 mA/cm² was achieved. With alternating current the optimum energy was achieved by using iron electrodes and was 0.69 kWh/m³ and 0.72 kg/m³ and with stainless steel electrodes were 0.98 kWh/m³ and 0.9 kg/m³. In direct current the optimum amount of energy with iron and stainless steel electrodes were respectively 1.97 kWh/m³ and 1.17 kg/m³ and 2.53 kWh/m³ and 1.42 kg/m³. The maximum amount of sludge was made in alternating current with a 0.084 kg/m³ current density and in direct current at 0.091 kg/m³. According to the results this process can be used as a suitable method for a wide use of electrolyte reactors in the industrial scale and for removing lead and zinc from aqueous environments.

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1. Introduction

Some industrial waste includes heavy metals such as cadmium, chromium, copper, nickel, mercury, lead and zinc which if drained directly into the environment are hazardous. Nowadays contamination with heavy metals is one of the most serious environmental disasters [1,2]. Drainage of heavy metal due to their dangerous side effects is a matter of concern in the environment. In contrast to organic pollutants, heavy metals are not biologically degradable, are highly soluble in water, have a tendency to accumulate in living organisms and many of their ions are toxic and carcinogenic. Lead and zinc are among the most toxic non necessary heavy metals in the environment and based on the severity of pollution are among the class one toxic pollutants [3,4]. Lead and zinc ions are high in waste water from different sources such as plating, lead

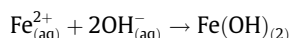
batteries, phosphate fertilizers, mines, dyes, refineries, paper and pulp industries. Lead and zinc toxication leads to kidney failure, lung fibrosis and cancer, also these ions have negative effects on blood and bones. The permissible amount of lead and zinc in drinking water is respectively 0.01 mg/l and 0.5 mg/l [5,6]. In order to prevent the negative effects of these metals on humans and the environment and also to follow the guidelines, it is necessary to remove these metals from wastewater before disposal. The routine methods for removing lead and zinc from water include ion exchange, reverse osmosis, floating, sequestration, coagulation, solvent extraction, chemical filtration, surface absorption and electrocoagulation. Each method has its own pros and cons. For example physical method such as ion exchange, reverse osmosis and electrodialysis for lead and zinc removal are expensive and inefficient. Chemical infiltration also has cons such as the high cost of maintenance, difficulty in transferring and disposing sludge and neutralizing the effluent, which is non-practical. Also removing lead and zinc from water by adsorption by different chemicals have

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been evaluated. The main cons of the experimented absorbents are their low efficiency and high cost [7–11]. From the different techniques mentioned above, one effective technology which fulfills the cleaning, applicability, investing costs, operation and environmental regulations is electro coagulation. This technology was used for the first time in the 19th century for the filtration of chips wastewater and recently is being used as a replacement for most popular filtration processes [12,13]. The electrocoagulation technologies are basically electrical processes and include unstabilizing suspending solids, emulsion or solved pollutants in aqueous solutions by using electrical currents [14,15]. Electrocoagulation (EC) is a process including hydro oxide metal flukes produced from wastewater by electrical dissolution of solvable anodes which are usually made from ferrous and aluminum. Due to electrochemical oxidation ferrous or aluminum is produced in the anode of metal cations, but in cathodes hydrogen production happens [16]. Despite this fact, it has to be considered that using Fe^{+3} as a coagulating factor in wastewater treatment has many advantages, because in comparison to the Al^{+3} ions which may have hazardous effects such as inducing Alzheimers disease, these ions are harmless [17]. Therefore in this study electrodes made from iron and stainless steel are used. In this process the main reactions that are used are as follows [18,19] Eqs. (1)–(4):

Anode:



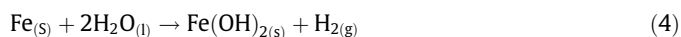
Cathode:



Chemical reaction:



In total:



The electrochemical process includes many physical and chemical processes such as evacuation anode oxidation, cathode reduction, coagulation, electrophoresis migration and surface absorption. Also during the electrocoagulation process in comparison to chemical sedimentation the fluid does not enrich with anions and the salt content does not increase. This process is referred to the production of metal sludge which by using electrocoagulation is more condensed than chemical sedimentation. Also, electrocoagulation needs simple equipment, has a high removal efficiency, has condensed treatment facilities, relatively low cost and the capability of completely automating the retention time is low and also its operation is easy. This characteristic is referred to its low operation costs for industrial use [20,21]. Electro coagulation has been used successfully for treating wastewater including electroplating wastewater, textile wastewater, olive oil factory wastewater, laundry wastewater, tannery wastewater, paper industry and pulp wastewater, brewing and slaughtering wastewater [22–25].

In this study the efficiency of electrocoagulation with alternating and direct current (AC & DC) by using iron alloy and stainless steel in removing lead and zinc has been evaluated. The effect of current density, energy consumption, initial pH, electrode quality, space between electrodes and the mixing rate on the efficiency of removal has been studied, in order to determine the optimum condition for operation.

2. Materials and methods

2.1. Wastewater characteristics

The wastewater used in this study was taken from the battery making industry wastewater at the Kerman industrial town and its characteristics have been shown in Table 1. Before this wastewater was used in the study, its suspended particles were separated by a filter.

2.2. Electrochemical cell

An electrochemical cell was made from plexiglass with a $12 \times 10 \times 12$ cm diameter. The efficient volume of the reactor was 1 l. A figure of this cell has been shown in Fig. 1. In every experiment the total volume of the wastewater was 1 l. Thirty Iron and stainless steel rods with 50 mm length and 5 mm diameter were used. Electrodes were distance of 2 cm from the bottom of the reactor. The electrodes were connected to each other serially and to the main source monopolar (only the two outer electrodes were connected to the positive and negative poles and the middle electrodes were connected together). The direct and alternating current was provided by one source model (GW GPC-3060D) and with a current density range of 2, 4, 6, 8 and 10 mA/cm^2 .

2.3. Experiments

The Wastewater samples were taken from the battery building industry and in less than 6 h were transferred to the laboratory. With samples were measured for lead and zinc by an atomic absorption model (UNICAM 924), electric conductivity with a conductivity meter (model 4071) and pH with a pH meter (model InoLab WTW), iron and sulfate by a spectrophotometer model AL800 based on the techniques mentioned in the book for standard techniques of water and wastewater experiment. Then the optimum pH for removing lead and zinc was determined and for keeping pH in the optimum range H_2SO_4 and NaOH , 0.1 N was used. The wastewater inside the reactor was mixed by a magnetic mixer. After each experiment, electrodes were washed with a brush and with diluted HCl. In order to watch the treatment progress, in time intervals of 10, 20, 30 and 40 min, 25 ml samples were taken from the middle of the reactor. Then the samples were passed through 0.45 μm filters in order to eliminate the floc. The filtered samples were stored at 4 °C and eventually were measured for lead and zinc by atomic absorption and according to the methods mention in the standard methods book for examining water and wastewater number 3111B. By the end of each study, the amount of producing sludge and the pH of the remaining solution were measured in order to find out about the amount of sediments and the changes in pH from this process. Also after each experiment, the electrodes were washed by diluting HCl solution and after washing again with distilled water, they were weighted accurately.

Table 1

The characteristic of the battery making industrial wastewater located at the Kerman Industrial Town.

Number	Wastewater characteristic	Amount
1	Lead (Pb)	9 mg/l
2	Zinc (Zn)	3.2 mg/l
3	Electrical conductivity	4.5 ms/cm
4	pH	2.8
5	Iron	2.3 mg/l
6	Sulfate	5 mg/l

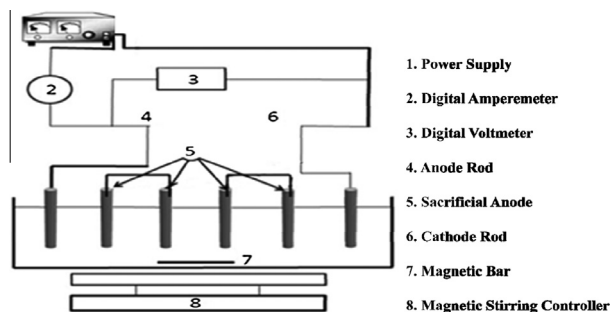


Fig. 1. Bench-scale EC reactor with monopolar electrodes in series connection.

2.4. Calculations

The amount of electrical energy used is an important economical parameter in the electrocoagulation process. This parameter is calculated by this equation (Eq. (5)):

$$E = \frac{U \cdot I \cdot t}{V} \quad (5)$$

E is the electrical energy used (in kW h/m³), U is the voltage used (in V), I is the current (in A), t is the coagulation time in hours. V is the volumes in liters. The amount of sacrificed electrode use is determined by measuring the initial and eventual weight of the electrodes.

3. Results

In the present study, electrocoagulation by iron rods was used as a treatment technology for removing lead and zinc from the battery making industrial wastewater and the efficiency of their removal by using direct and alternating current was evaluated. The results of this study have been shown in Tables 2 and 3 and Figs. 2–12. In Fig. 2, the best pH for removing lead and zinc has been shown which was 5 (removal efficiency 98.4%) and 4 (removal efficiency 98.2%) respectively. In Tables 2 and 3 the efficiency of removing lead and zinc with iron and stainless steel rod electrodes and by using alternating and direct current in the different contact times has been shown. As these tables show by using alternating current the current density of optimum in removing lead and zinc by iron rod electrodes was achieved in 6 mA/cm² and in a reaction time of 30 min and was respectively 96.7% and 95.2%, and with a stainless steel rod electrode was achieved in optimum current density of 8 mA/cm² and reaction time of 40 min respectively was 93.8% and 93.3%. By using direct current the removal efficiency of lead and zinc with iron rod electrodes was respectively 97.2% and 95.5% in optimum current density of 6 mA/cm² and with stainless steel rod electrodes was 93.2% and 92.5% in optimum current density of 8 mA/cm² and 40 min time.

In Figs. 3 and 4 the energy consumption of both alternating and direct current with iron and stainless steel rod electrodes has been shown. With alternating current the maximum energy consumption was by using iron rod electrodes in optimum current density of lead and zinc removal which was 0.69 and 0.69 kW h/m³ and with stainless steel rod electrodes was 0.89 and 0.96 kW h/m³. In direct current the highest amount of energy consumption was with iron rod electrodes in optimum current density of lead and zinc removal which was 1.93 and 1.97 kW h/m³ and with stainless steel was 2.94 and 2.53. Also the amount of electrode used in alternating current by using iron rod electrodes in optimum current density of removing lead and zinc was respectively 0.69 and 0.72 kg/m³ and with stainless steel rod electrodes was 0.87 and 0.9 kg/m³, and in direct current with iron rod electrodes in optimum current density

current in lead removal was 1.13 kg/m³ and in zinc removal was 1.17 kg/m³ and with stainless steel rod electrodes was 1.35 kg/m³ and 1.42 (Figs. 5 and 6). The amount of sludge produced in this process has been shown in Figs. 7 and 8. As it can be seen in alternating current by using iron rod electrodes for removing lead and zinc in optimum current density current the produced sludge was 0.081 kg/m³ and 0.084 and with stainless steel was 0.069 kg/m³ and 0.073. In direct current by using iron rod electrodes the produced sludge in optimum current density for lead removal was 0.088 kg/m³ and for zinc removal was 0.091 kg/m³ and with steel electrodes was 0.079 and 0.081 kg/m³ respectively. The effect of the distance between the electrodes with lead and zinc removal was also studied and its effect has been shown in Figs. 9 and 10. In alternating and direct current the highest efficiency was achieved in 1 cm electrode distance. In Figs. 11 and 12 the pH change after the end of each stage of the experiment has been shown. The most change in pH was achieved by using iron rod electrodes in optimum current density of lead and zinc removal and was respectively 1.44, 1.12 and with stainless steel rod electrodes was 1.62 and 0.79. In direct current the largest pH changes were by using iron rod electrodes in removing lead and zinc and in optimum current density and was respectively 1.89 and 1.33 and by using stainless steel rod electrodes were 1.38 and 0.98.

4. Discussion

In the present study, different parameters such as current density, pH, reaction time, amount of energy, the electrodes used type of electrode, electrode design, and distance between electrodes on the efficiency of the electrocoagulation process in removal of heavy metals such as lead and zinc and the amount of sludge produced in this process have been evaluated.

4.1. The effect of current density on process efficiency

In all of the electrochemical processes the current density is the most important parameter for controlling the reaction rate inside the electrochemical reactor. It has been well shown that the current density can determine the production rate of coagulant, the production rate of bubbles, its size and distributions and the rate of floc growth in electrocoagulation reaction with different electrodes [26,27]. The effect of current density on the removal of lead and zinc from the battery making industry wastewater by two types of current (alternating and direct) and two types of electrode (iron and stainless steel) were evaluated. From the results (Tables 2 and 3), it is found that, as current density increases, removal of lead and zinc also increases. In this study, in order to reach an appropriate removal efficiency of lead and zinc optimum current density for both alternating and direct current with iron rod electrodes was obtained 6 mA/cm² and with stainless steel rod electrodes was 8 mA/cm². The results showed that alternating current and iron rod electrodes are relatively more effective than direct current and stainless steel rod electrodes. This is probably due to the uniform decomposition of the anode and cathode in alternating current and during the time of electrocoagulation. The removal efficiency shows that by increase in the current density the removal of lead and zinc increase. This can be attributed to the increase in the amount of absorbent [Fe(OH)₃] which are produced in situ thereby resulting in rapid removal of lead and zinc. The amount of absorbents is determined by the Faraday Law [26,28,29] (Eq. (6)):

$$C = \frac{I \cdot t \cdot M}{Z \cdot F \cdot V} \quad (6)$$

In which C is the current density of iron in the electrolytic cell in kg/m², I is the current in A, t is the duration of electrocoagulation in

Table 2

The efficiency of lead removal (percent) by using iron and stainless steel rod electrodes and alternating and direct current.

Electrode and current type	Contact time (min)	Current density (mA/cm ²)					Electrode and current type	Contact time (min)	Current density (mA/cm ²)				
		2	4	6	8	10			2	4	6	8	10
Iron rod electrode, alternating current	10	67.6	74.5	79.4	83.8	86.4	Iron rod electrode, direct current	10	65.9	72.6	77.5	81.8	85.4
	20	78.5	85.3	90.2	95.6	96.7		20	76.7	84.2	89.3	93.7	96.6
	30	87.4	89.7	96.7	97.4	98		30	87.1	87.8	95.2	96.3	97.2
	40	89.7	92.2	97.4	98.2	98.4		40	88.9	91.4	97.2	97.5	98
Stainless steel rod electrode, alternating current	10	56.4	62.6	68.7	71.6	75.4	Stainless steel rod electrode, direct current	10	54.8	60.7	66.4	70.3	73.4
	20	64.7	75.4	79.2	83.7	89.3		20	63.5	75.2	77.8	82.6	88.6
	30	77.3	84.7	88.3	91.4	92.6		30	75.8	83.4	87.4	91.1	91.3
	40	84.6	89.2	91.4	93.8	95.3		40	84.2	87.9	90.8	93.2	94.8

Table 3

The efficiency of zinc removal (percent) by using iron and stainless steel rod electrodes and alternating and direct current.

Electrode and current type	Contact time (min)	Current density (mA/cm ²)					Electrode and current type	Contact time (min)	Current density (mA/cm ²)				
		2	4	6	8	10			2	4	6	8	10
Iron electrode, alternative current	10	69.2	75.8	77.6	84.7	86.5	Iron electrode, direct current	10	67.4	73.8	75.6	82.7	85.3
	20	76.5	87.5	89.2	95.3	95.9		20	75.9	86.4	87.4	93.5	93.9
	30	88.4	91.4	95.2	96.4	97.3		30	87.6	91.2	93.7	95.8	96.5
	40	90.7	93.3	96.5	97.8	98.2		40	89.5	92.7	95.5	97.3	97.7
Stainless steel electrode, alternative current	10	55.3	63.6	66.8	73.7	78.3	Stainless steel electrode, direct current	10	54.4	62.5	66.3	71.8	76.7
	20	63.7	72.8	77.5	85.4	87.7		20	61.8	70.9	75.6	84.2	86.9
	30	77.5	86.3	89.3	92.7	94.7		30	75.6	85.7	88.5	91.3	93.6
	40	85.8	89.7	92.6	93.3	95.8		40	85.2	88.5	90.7	92.5	95.3

seconds, M is the anode molecular mass in kg/mol which is 0.05585 for iron, zinc is the chemical equivalent (for iron = 2), F is the Faraday constant rate (26,485 C/mol) and V is the sample volume in m³. As it was expected the amount of surface lead and zinc absorption increases which increase in absorbent current density which it is directly related to the current density and reaction time and shows that surface absorption is highly related to the accessibility of positions for lead and zinc to bind [30,31]. The decrease in lead and zinc is more with iron rod electrodes than stainless steel rod electrodes and this fact is mainly due to the quicker formation of Ferric sulfate and hydroxide ferric from usual iron, which has less resistance to rusting in comparison to stainless steel [32]. As the current density of current increases, the amount of lead and zinc decrease more. This event is mainly due to the fact that increase in current density leads to bigger oxygen bubbles produced by the anode which itself leads to upward current and quicker lead and zinc removal by floatation. Meanwhile, the main effect of increased current is more degradation of the anode which leads to increase in the concentration of metal ions in the solution and therefore the rate of lead and zinc removal increases [18]. Also as it can be seen in the figure, with increasing current density the current needed for lead and zinc removal decreases. These results are in line with the results from Yilmaz et al. in separating out of warm subsurface waters by electrocoagulation, Ghosh et al. in removing iron (II) from tap water by using electrocoagulation techniques and Merzouk et al. in removing turbidity and separating heavy metals by using electrocoagulation techniques and floatation [27,33,34].

4.2. The effect of initial pH

In chemical or electrochemical segregation processes, pH is an important parameter and has a significant effect on the formation of hydroxide metal types and the mechanism of ion and pollutant removal [21,35]. The effect of pH on alternating and direct current by using iron and stainless steel rod electrodes in the removal of lead and zinc from wastewater were evaluated. The removal efficiency of lead and zinc was increased with increasing the pH and the maximum removal efficiency of 98.4% and 98.2% were obtained

at pH 5 and 4 respectively. The minimum removal efficiency of lead was 54.6% and zinc was 66.4% at pH 3. The lead and zinc removal efficiencies decreased in lower acidic and higher alkaline pH values since hydroxide ions were oxidized at the anode as pH was greater than 5 for lead and 4 for zinc. In addition, $\text{Fe}(\text{OH})_6^{3-}$ and $\text{Fe}(\text{OH})_4^-$ ions may be present at high pH, which lacks a removing capacity. At lower pH the protons in the solution were reduced to H_2 at the cathode and the same proportion of hydroxide ions could not be produced [18]. Also at more acidic and alkaline pH show to an amphoteric behavior of $\text{Fe}(\text{OH})_3$ which leads to soluble Fe^{2+} cations (at acidic pH) and to monomeric anions $\text{Fe}(\text{OH})_4^-$ (at alkaline pH) and therefore metal removal decreases. It is well known that these soluble species are not useful for water and wastewater treatment [31]. Usually the pH changes during the process time of EC and this change depends on the type of electrode and initial pH. Increase in pH in this process is thought to be related to the formation of H_2 gas in the cathode electrode and accumulation of hydroxide ions in the solution. Therefore the electrocoagulation process can function as a stabilizer for pH or in other words the EC process shows a buffering potential, because it balances the production and consumption of OH^- and this prevents the changes in pH [36,37]. These results are in line with the study done by Escobar et al. in optimizing the electrocoagulation process for removing copper, lead and cadmium from natural waters and synthetic wastewater, Heidmann et al. which studies the removal of zinc, copper, nickel, mercury and chromium in aqueous solutions with electrocoagulation and Canizares et al. which studies the effect of pH as an important parameter in choosing coagulation and electrocoagulation for treating wastewater [38–40]. The pH of the solution increases during electrolysis, but in very acidious electrolytes, the bases produced during the electrolysis process is not enough for increasing the pH of the solution [41]. The pH of the samples by the end of the measurement process and its changes has been shown in Figs. 9 and 10. The biggest changes in pH were seen in alternating current and by using iron rod electrodes in optimum current density for removal of lead and zinc were 1.44, 1.12 and with stainless steel rod electrodes were 1.62 and 0.79. In direct current the largest changes in pH

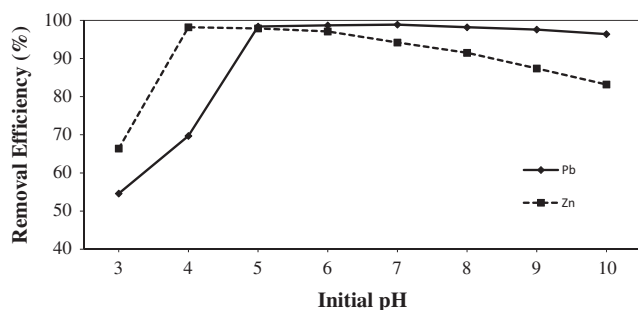


Fig. 2. Determining the optimum pH of removing lead and zinc with direct and alternating current and by using iron and stainless steel electrodes (current density 4 mA/cm², contact time 20 min, distance between electrodes 1.5 cm, mixing rate 200 rpm).

were seen by using iron rod electrodes and in removing lead and zinc in optimum current density which was 1.89, 1.33 respectively and by using stainless steel rod electrodes was 1.38 and 0.98 respectively.

4.3. The effect of reaction time

In the electrocoagulation process the time of electrolysis affects the treatment efficiency. Electrocoagulation experiments were performed in 40 min with different current densities and different pHs. Tables 2 and 3 show the relation between the reaction time and the efficiency of lead and zinc removal in different current

densities. As it can be seen in these tables, as the reaction times increases, the removal efficiency increases. This result is thought to be related to this fact that the efficiency of the process is directly related to the concentration of hydroxide and metal ions that are produced by the electrodes and as time passes the ion concentration and hydroxide clots increase [20–42]. The reaction time determines the production of Fe³⁺ ions from the iron and stainless steel rod electrodes. During the time of electrolysis the positive electrodes undergo anode reactions. The released ions neutralize the electrical load of the pollutant and therefore coagulation starts. When the time for electrolysis increases, the concentration of the ions and hydroxide clots increase. Also in high current, more bubbles are produced and lead to better mixing of metal hydroxides and pollutants and eventually better floatation capability and increased removal efficiency [16–43]. In this study the optimum time for lead and zinc removal in alternating current by using iron and stainless steel rod electrodes was 30 and 40 min and in direct current with both iron and stainless steel rod electrodes was 40 min. This result was similar to the results from the Drouiche et al. study in evaluating the treatment of photovoltaic wastewater by electro coagulation and removal of fluoride by aluminum electrodes and Mouedhen et al.'s study in evaluating the behavior of aluminum electrodes in the electrocoagulation process [16–41].

4.4. The amount of energy and electrode used

The amount of energy used is an important economic parameter in the electrocoagulation process. The majority of operating

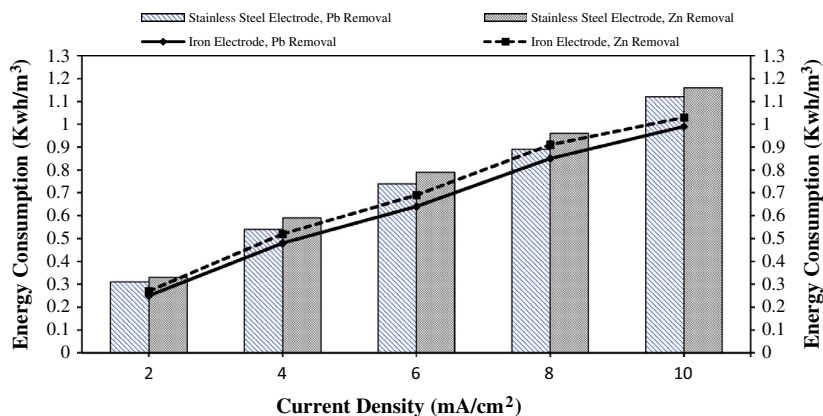


Fig. 3. The amount of energy consumption in removing lead and zinc by using alternating current (current density 6 mA/cm², pH 5 for removal of Pb and 4 for removal Zn, contact time 30 min, distance between electrodes 1.5 cm, mixing rate 200 rpm).

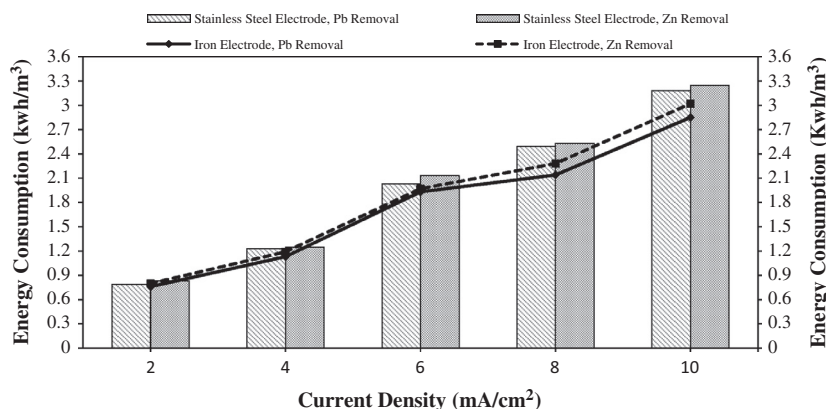


Fig. 4. The amount of energy consumption in removing lead and zinc by using direct current (current density 6 mA/cm², pH 5 for removal of Pb and 4 for removal Zn, contact time 30 min, distance between electrodes 1.5 cm, mixing rate 200 rpm).

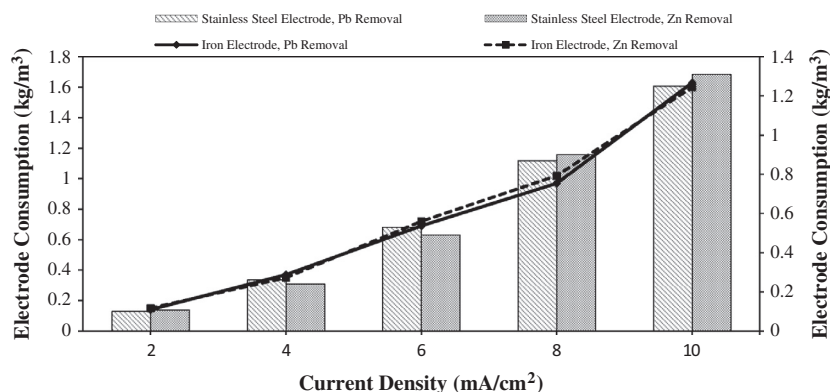


Fig. 5. The amount of electrode used in removing lead and zinc by using alternating current (current density 6 mA/cm², pH 5 for removal of Pb and 4 for removal Zn, contact time 30 min, distance between electrodes 1.5 cm, mixing rate 200 rpm).

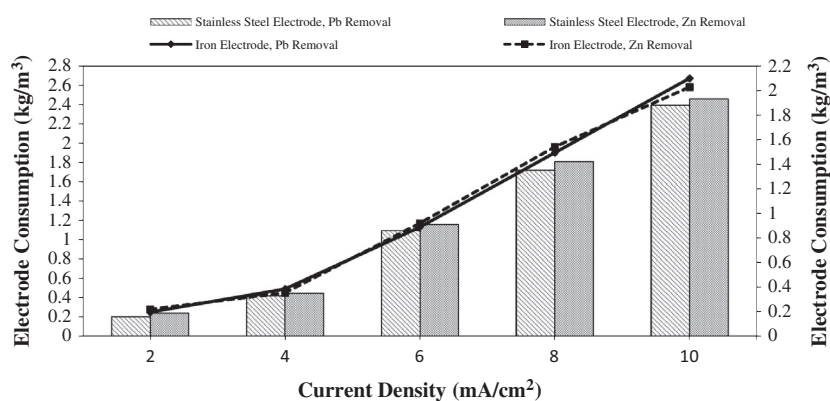


Fig. 6. The amount of electrode used in removing lead and zinc by using direct current (current density 6 mA/cm², pH 5 for removal of Pb and 4 for removal Zn, contact time 30 min, distance between electrodes 1.5 cm, mixing rate 200 rpm).

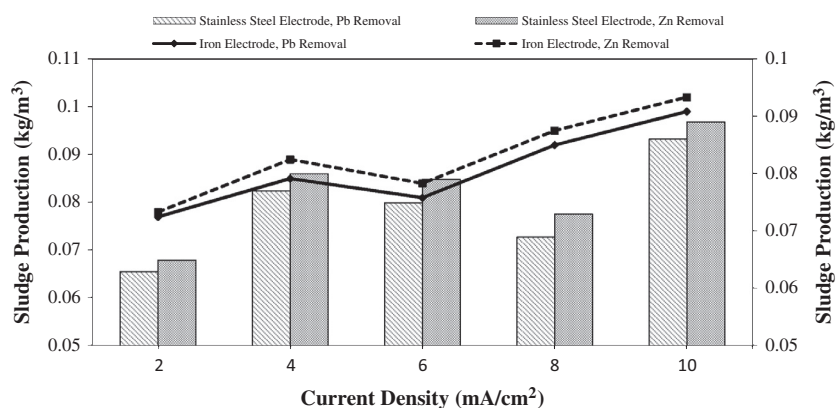


Fig. 7. The amount of sludge produced in alternating current by using iron and stainless steel electrodes (current density 6 mA/cm², pH 5 for removal of Pb and 4 for removal Zn, contact time 30 min, distance between electrodes 1.5 cm, mixing rate 200 rpm).

costs is also related to the amount of electrical energy used. Also in this study the amount of energy used for lead and zinc removal was evaluated as a function of time and used electrodes in different current densities. When the current density increases from 2 to 6, the energy consumption in alternating and direct current with iron and stainless steel rod electrodes increased slightly. When the current density increased from 6 to 10 with alternating current, energy consumption slightly increased and with direct current increased significantly. Results showed that increase in current leads to increase in energy consumption due to polarization and

increase in the number of iron and stainless steel compounds, which elevate the efficiency of lead and zinc removal as well. Therefore in order to reach an optimum current density, it's necessary to evaluate both the removal percent and the amount of electrical energy used [23–44]. Energy consumption versus current density for both alternating and direct current with both electrodes was shown in Figs. 3 and 4. With alternating current the maximum energy consumption was by using iron rod electrodes in optimum current density of lead and zinc removal which was 0.69 and 0.69 kW h/m³ and with stainless steel rod electrodes was 0.89

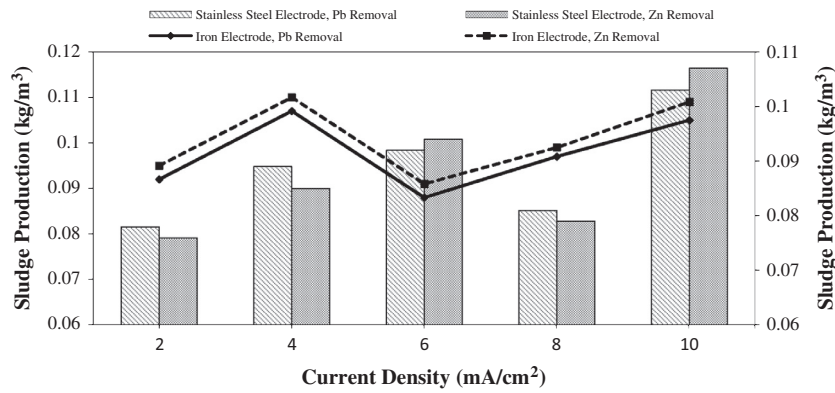


Fig. 8. The amount of sludge produced in direct current by using iron and stainless steel electrodes (current density 6 mA/cm², pH 5 for removal of Pb and 4 for removal Zn, contact time 30 min, distance between electrodes 1.5 cm, mixing rate 200 rpm).

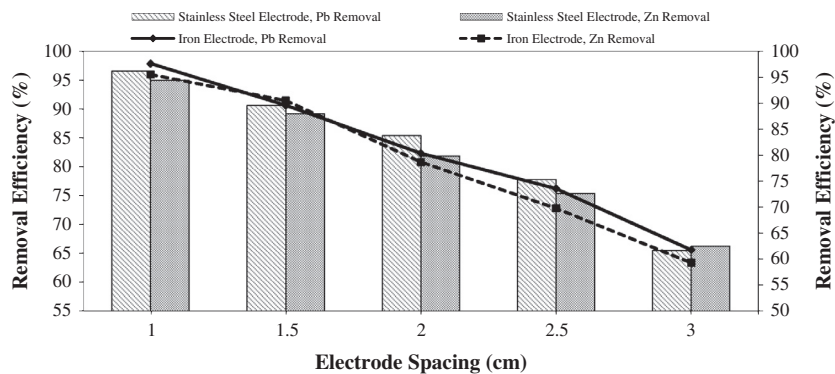


Fig. 9. The efficiency of lead and zinc removal by using alternating current in different distances between electrodes (current density 6 mA/cm², pH 5 for removal of Pb and 4 for removal Zn, contact time 30 min, mixing rate 200 rpm).

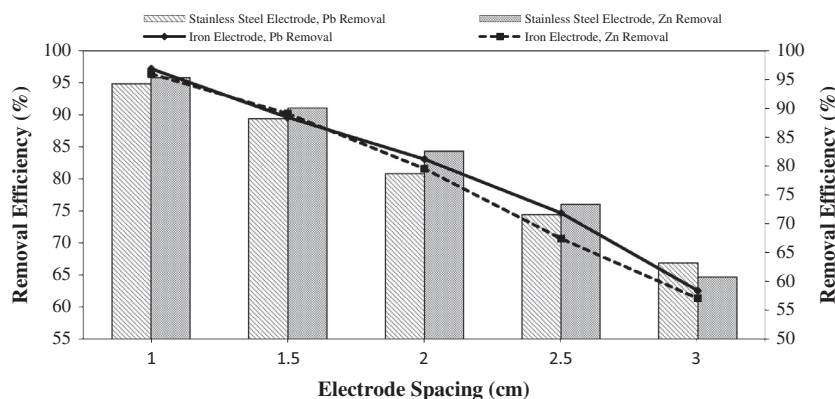


Fig. 10. The efficiency of lead and zinc removal by using direct current in different distances between the electrodes (current density 6 mA/cm², pH 5 for removal of Pb and 4 for removal Zn, contact time 30 min, mixing rate 200 rpm).

and 0.96 kW h/m³. In direct current the highest amount of energy consumption was with iron rod electrodes in optimum current density of lead and zinc removal which was 1.93 and 1.97 kW h/m³ and with stainless steel rod electrodes was 2.94 and 2.53. Therefore energy consumption by using iron rod electrodes was less than with stainless steel rod electrodes, and was probably because of the high voltage that was necessary for reaching the optimum current density in the serial design model. The high current density also leads to more decrease in the weight of the electrodes and an increase in the amount of electrodes released into the solution [26–33]. The amount of electrode used in the different current

densities has been shown in Figs. 5 and 6. The amount of iron rod electrodes used in optimum current density for lead and zinc removal by using alternating current was respectively 0.69 and 0.72 kg/m³ and the amount of stainless steel rod electrode used was 0.87 and 0.9 kg/m³. By using direct current, the amount of iron rod electrode used in the optimum current density for lead and zinc removal was 1.13 kg/m³ and 1.17 and the amount of stainless steel rod electrode used was 1.35 kg/m³ and 1.42 respectively. These results were inline with the results from the Zared et al. study in electrocoagulation treatment of the black liquid from the paper making industry and Sengil et al. in treating tannery

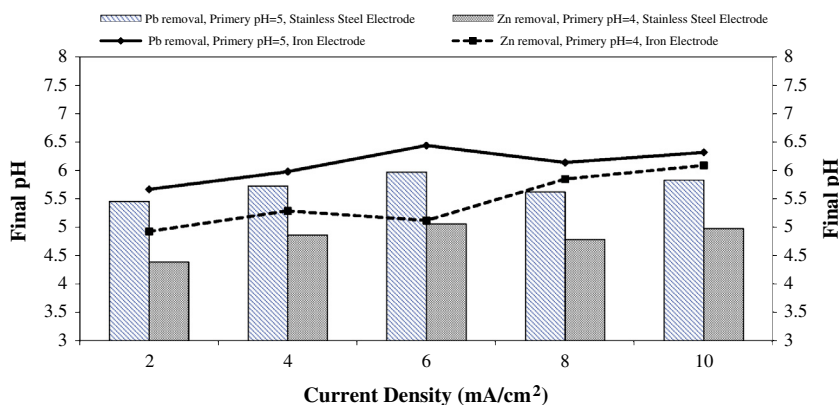


Fig. 11. The amount of initial pH change in lead and zinc in removal by using alternating current.

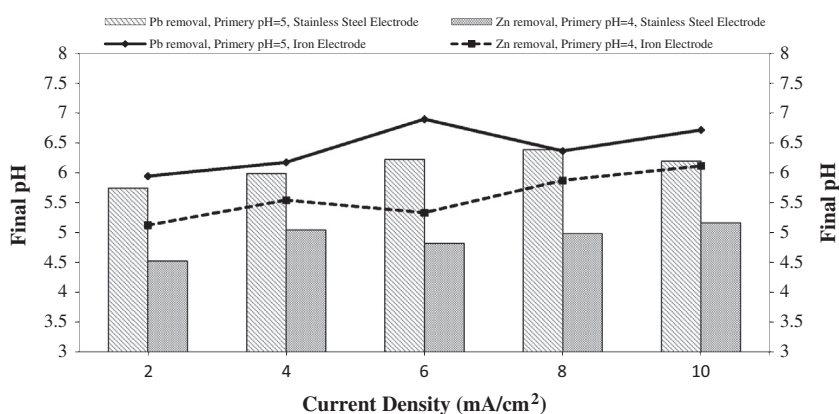


Fig. 12. The amount of initial pH change in lead and zinc in removal by using direct current.

wastewater by electrocoagulation and Chou et al. in electrochemical removal of Indium ions from aqueous solutions by using iron rod electrodes [23–45].

4.5. The effect of electrode type

The electrode of electrochemical cells is the heart of the reactor. Therefore choosing the right material is very important. The most usual material used for electrodes in electrocoagulation is iron, aluminum and stainless steel. These materials are cheap, accessible and efficient [12]. Therefore in this study two iron and stainless steel rod electrodes in similar experimental conditions were evaluated. The results for lead and zinc removal for these two types of electrodes by using different current densities have been shown in Tables 2 and 3. As it can be seen an almost similar efficiency in decreasing lead and zinc was seen in both electrodes. As time passes the amount of removal by iron rod electrodes increases slightly in comparison to stainless steel rod electrodes. The sample of treated water with iron and stainless steel rod electrodes first turned green and then became brown and turbid. Green and yellow color can be due to the production of iron 2 and 3 ions during the time of electrolysis which suspend in the remaining solution. Also they can eliminate the lead and zinc ions by combination or by electrostatic absorption after coagulation. Similar studies by Chen et al. during removal of oil and grease from the wastewater of restaurants by electrocoagulation and Zaied et al. in coagulation treatment of black water from the paper making industry has been done. They described the color change and noted that the color change was from gradual oxidation of Ferrous (II) ions produced

by the electrical breakdown in the anode in the presence of dissolved oxygen [45,46]. On the other side based on the amount of electrical energy used and the sacrificed electrode which affects the cost of the process, the iron rod electrode was more efficient than the stainless steel rod electrode [12]. The ferrihydroxy compounds are produced after initial oxidation at the anode and the most dominant from of it $\text{Fe}(\text{OH})_2$ is non soluble in a wide range of pHs. The concentration of iron in water play an important role in the removal of pollutants, and determines the concentration and pH of different probable chemical types of iron in the aqueous solution [47]. The effect of the released hydrogen at the cathode is to facilitate the progress of floc accumulation on the surface of the solution in the shape of a thick whitish mud. One of the mechanisms that has been suggested in relation to the production of hydroxide ions has been mentioned here [46] Eqs. (7)–(13):

$\text{PH} < 4$

Anode:



Cathode:

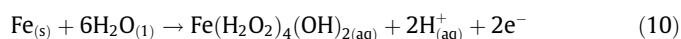


Overall:



$4 < \text{PH} < 9$

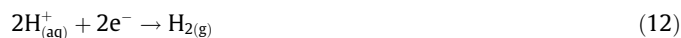
Anode:



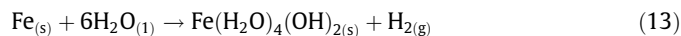
Bulk of solution:



Cathode:



Overall:



4.6. The effect of electrodes design

In every electrochemical process different types of electrodes and the type of paired electrodes are known as effective factors on the process performance. Therefore, it is important to choose the right pair of electrodes. In this study for determining the optimum pair of electrodes, the iron and stainless steel rod electrodes in two different complexes of iron–iron and steel–steel in similar conditions were evaluated. Tables 2 and 3 show the effect of different pairs of electrodes in the efficiency of lead and zinc removal. As it can be seen in the figures, high removal efficiency was achieved with iron–iron pair electrodes. In alternating current for lead 96.7% and for zinc 95.2% in optimum current density 6 mA/cm² and optimum reaction time of 30 min was achieved and in direct current of lead and zinc respectively 97.2% and 95.5% in optimum current density of 6 mA/cm² and optimum duration time of 40 min was achieved. This high efficiency in iron–iron pair electrodes is attributed to the chemical reaction which happens in the anode (Eq. (14)):



In the initial phase, iron ions are very efficient coagulants for particulate clotting. From the above reaction the electrochemical mass equivalent for Fe can be calculated. Therefore the electrochemical mass equivalent of iron is 1041 mg/Ah and this makes more coagulants to be theoretically produced from the iron anodes when a similar electrical load passes. This is probably a reason for higher removal efficiency of the iron rod electrodes [48]. Also the higher efficiency of the iron–iron current density can be attributed to the higher gradient potential of electrode and therefore more oxidation potential between anode and cathode in comparison to other designs. This leads to more coagulant production and better removal of the pollutants [32]. These results show that a reliable electrode design can be more efficient and economic in practice. Contrary to this El-Naas et al. in the treatment of refinery wastewater found a better operation for aluminum in comparison to iron as an electrode in the electrocoagulation process [32]. Therefore the better design and material for the anode deeply depends on the type of pollutants and the operation condition. In the electrocoagulation cell by designing the electrode the reaction will happens as follows [49] Eqs. (15)–(20):

Anodic reactions:

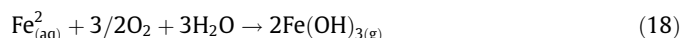


Cathodic reaction:

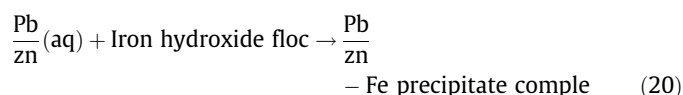


Liquid bulk reactions:

(6 < PH < 10)



PbZn removal



Therefore the main mechanism in removing lead and zinc from wastewater by the electrocoagulation process may include Fe oxidation to ferrous ions and simultaneous electrolysis of anode on the anode surface which leads to the production of oxygen, oxidation of ferrous ions to ferric by reacting with oxygen molecules and formation of hydroxide, poly hydroxide, and precipitating polyferrous hydroxide [48,49].

4.7. The distance between electrodes

The effect of the distance between electrodes for better pollutant removal was evaluated. When the distance between the electrodes increases, the electrical current decreases and for reaching a certain current density the voltage has to increase [29–32]. Removal percentages for lead and zinc with various intervals between electrodes by using alternating and direct current were investigated and shown in Figs. 9 and 10. It has been observed maximum removal efficiency for both current (AC & DC) with both electrodes was achieved at a distance of 1 cm and by increasing the inter-electrode intervals, the removal percentage of lead and zinc decreases. In this distance EC efficiency by using alternating current with iron rod electrodes for lead and zinc removal was 97.8% and 95.9% and with stainless steel rod electrodes was 96.1% and 94.3% respectively. In direct current in distance of 1 cm between electrodes the maximum removal efficiency lead and zinc with iron rod electrodes was 97.2% and 96.4% and with stainless steel rod electrodes was 94.3% and 95.3% respectively. This high efficiency probably happens due to the electrostatic effects related to the distance between the electrodes. Therefore when this distance increases, the electro statistic effect decreases, the mobility of the produced ions decreases and there is more time for accumulation and clot production. In other words, the reason for efficiency decrease with distance increase is local concentration decrease and electrostatic attraction which itself can lead to slower substitution of produced ions during the time of electrolysis and easier clot production [42]. In the similar studies by Nanseu-Njiki et al. for removing mercury from water by electrocoagulation, El-Naas et al. in evaluating electrocoagulation for treating the wastewater from oil refinery and Modirshahla et al. in nitrophenol removal from aqueous solutions by electrocoagulation a similar behavior was seen [32–46]. Also in electrochemical anode oxidation by using voltage for initial electrodes and as time continues a very thin layer of metal hydroxide forms on the anode which leads to external resistance. This resistance increases as the distance between electrodes increases. Therefore after some time the operation current decreases and for stabilizing the current voltage should increase, but this leads to increase in resistance. There for ohmic potential increases and at the beginning prevents anode oxidation. As the rate of anode oxidation decreases, the number of cations in the anode also decreases. These cations are responsible for coagulant formation. Therefore by increase in the distance between electrodes from 10 mm, the amount of particulate accumulation and surface absorption of pollutants decreases as well. This may lead to low efficiency in big distances between electrodes. In minimum distance between electrodes resistance is less which facilitates the electrolysis process and leads to better lead and zinc removal [33].

4.8. The effect of mixing rate

The electrocoagulation process is an electrochemical treatment process in which is achieved by the movement of ions by using electricity. During this process a stable solution environment is very efficient for the movement of ions. Mixing the environment leads to chaos in the movement of ions and waste of electricity [50]. High mixing speeds places the ions on the way of the electrodes and leads to more product formation and on sufficient separation of salt sediments. Therefore in this study for reaching a complex but omissible sediment 200 rpm was chosen as optimal mixing speed. The percent of lead and zinc removal increases by increasing mixing speed up to 200 rpm significantly which is due to the formation and sticking of flocs together and therefore easier sedimentation. In mixing speeds more than 200 external mass transfer is very small or negligible [36–42]. In contrast to this, in Sirajuddin et al.'s study in electrical retrieval of chromium salts from tannery wastewater, the mixing speed of 500 rpm was the optimum [51]. But in Ilhan et al.'s study by treating latex with electrocoagulation by aluminum and iron rod electrodes the speed of 200 rpm was determined as optimum [50]. Also by using rod electrodes instead of flat ones the possibility of placing more electrodes in the reactor space was provided. On the other side, the mixing process in comparison to the situation where flat electrodes were placed by each other was done better and led to the production of more metal hydroxide clot in a certain time, shorter process time and less operation problems [17–52].

4.9. The amount of sludge produced from electrocoagulation

Sludge production is one of the important parameters in describing the electrocoagulation process. The amount of sludge produced during EC is related to the treatment industry. This factor is also related to the problem of production and disposal of solid waste from this process. In this process, many pollutants are separated and float on the surface of wastewater as sludge. The sludge produced from electrocoagulation is related to the characteristics of crude wastewater, disposable solids and unstabilized material through coagulation and the coagulant concentration and is also related to the current density and the retaining time [26]. The amount of sludge produced in this study has been shown in Figs. 7 and 8. The amount of sludge produced increases with increase in lead and zinc removal efficiency from wastewater. As the current density increases, anode degradation happens appropriately and the remaining of metal sludge increases and the lead and zinc ions are eliminated by sedimentation or adsorption on metal sludge efficiently. The amount of sludge in this process in both direct and alternating current is achieved by using iron and stainless steel rod electrodes. In alternating current to iron rod electrodes, the highest amount of sludge was produced in optimum current density of lead and zinc, 0.081 kg/m³ and 0.084 and by steel electrodes in 0.069 kg/m³ and 0.073. The highest amount of sludge was produced in direct current by using iron and stainless steel rod electrodes in optimum current density for lead removal at 0.081 and 0.088 kg/m³ and zinc removal of 0.091 kg/m³ and 0.079. It seems like the hydroxide iron links with water are chemically and physically more than the hydroxide steel links [25–28].

5. Conclusion

In the present study have been tried to evaluate the capacity of using the electrocoagulation process in treating the battery making wastewater. The effect of variables such as current density, electrode type, design and distance between electrodes, electrolysis time and wastewater pH on the removal of lead and zinc was also

evaluated. The results showed that electrocoagulation can effectively reduce the high amounts of metal ions by hydroxide iron and steel flocs as absorbents. Lead and zinc were eliminated by chemical sedimentation by hydroxide ions formed at the cathode by water electrolysis and by co-sedimentation by iron and steel hydroxides. The removal efficiency increases by an increase in current density due to increase in the formation rate of hydroxide and steel flocs. Results showed that optimum removal of lead and zinc in current density of 6 and 8 mA/cm² was achieved which was respectively 96.7%, 95.2%, 93.8% and 93.3%. In direct current the optimum removal of lead and zinc by iron rod electrodes were respectively 97.2%, 95.5% and with stainless steel rod electrodes was 93.2% and 92.5% in current density of 8 and 6 mA/cm². The optimum electrolysis time with iron rod electrodes was 30 min and with stainless steel rod electrodes was 40 min. The least operation cost from the viewpoint of energy consumption and electrodes was achieved in alternating current. Also in alternating current less sludge than direct current was produced.

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